

That is, it is respectfully submitted that the teachings of the references as applied by the Examiner would have neither disclosed nor would have suggested such a non-aqueous electrolyte secondary battery negative electrode material, or the non-aqueous electrolyte secondary battery negative electrode using such material, or the non-aqueous electrolyte secondary battery using such negative electrode using such material, as in the present claims, including, inter alia, wherein each of the graphite particles included in the negative electrode has a structure where a plurality of flat graphite fine particulate assemblies or bonds non-parallel with each other, and each of the graphite particles has a layer of carbon formed on the surface of such graphite particle, with a ratio (by weight ratio) of the layer of carbon to a respective graphite particle being in the range of 0.001-0.01, among other characteristics of the graphite particles, and unexpectedly better results achieved in excellent discharge capacity, charge/discharge efficiency and charge load characteristics, where the graphite particle is covered with a layer of carbon with a weight ratio of layer of carbon to respective graphite particle as in the present claims. See claim 1.

It is respectfully submitted that the evidence already of record, in Applicants' specification and the evidence in the enclosed Declaration, establishes the unexpectedly better results achieved by the presently claimed negative electrode material having both excellent charge load characteristics and excellent bulk density variation rate when pressure is released.

In addition to the evidence in Applicants' specification, Applicants are filing further evidence for establishing unexpectedly better results. Attention is respectfully directed to the attached Declaration, including Table 1 on page 7 thereof, showing properties of the negative electrode material for Examples 1-4 and Comparative Examples 1-3 in Applicants' specification, and two (2) "Experiments" (Experiment 1

and Experiment 2) having superficial carbon rates just above the maximum superficial carbon rate in the present claims. That is, the Experiments 1 and 2 have superficial carbon rates of 0.015 and 0.02, both outside the scope of the present claims. Note that these Experiments 1 and 2, while having charge load characteristics that are good, have bulk density variation rates when pressure is released that is insufficient. It is respectfully submitted that only where the weight ratio of the layer of carbon to the respective graphite particle is in the range as in the present claims, are satisfactory properties, including bulk density variation rate when pressure is released, and charge/discharge characteristics, achieved. See the Results on page 8 of the enclosed Declaration.

Note that the Experiments in the enclosed Declaration were performed with procedures like the Examples described in the specification of the above-identified application (see Item 2 on pages 2-6 of the enclosed Declaration, and Table 2 on page 8 thereof).

Note also paragraph [0018] bridging pages 8 and 9 of Applicants' specification, describing that when the ratio of the superficial carbon is less than 0.001, an improvement width in the charge load characteristics is small, while when the ratio of the superficial carbon exceeds 0.01, the initial charge/discharge efficiency is deteriorated. Satisfactory properties occur with a carbon layer ratio as in all of the present claims.

In Item 3 on page 2 of the Office Action mailed April 28, 2010, the Examiner contends that Tables 2 (page 29) and 5 (page 34) in Applicants' specification "only have one data point outside of the claimed range and appear to show that the bulk density variation scales with the superficial carbon rate". It is respectfully submitted that the Examiner errs in contending that tables 2 and 5 "only have one data point outside of the claimed range. Thus, it is respectfully submitted

that each of Comparative Examples 1-3 in Table 2 are outside of the claimed range, Comparative Example 3 being just below the minimum of the claimed range and Comparative Example 2 being just above the maximum of the claimed range". In Table 5 on page 34 of Applicants' specification, each of Comparative Examples 4 and 5 have superficial carbon rates outside the scope of the present claims, with Comparative Example 5 being just below the minimum and Comparative Example 4 being just above the maximum. Note also Experiments 1 and 2 of the enclosed Declaration, having a superficial carbon rate above the maximum for the range in claim 1. Thus, it is respectfully submitted that each of Comparative Examples 1-5, and Experiments 1 and 2, has have data points outside of the claimed range with respect to superficial carbon rate, and note in particular Comparative Examples 2-5 and Experiments 1 and 2 have superficial carbon rates, relatively speaking, just outside the claimed range. It is respectfully submitted that this evidence is clearly appropriate for showing unexpectedly better results for the claimed range. See Manual of Patent Examining Procedure (MPEP) 716.02 (d), § II.

Properly considering all of the evidence of record in Applicants' specification as required under MPEP 716.01 (a) ("Examiners must consider comparative data in the specification which is intended to illustrate the claimed invention in reaching a conclusion with regard to the obviousness of the claims."), it is respectfully submitted that this evidence shows unexpectedly advantageous results, in, inter alia, bulk density variation rate when pressure is released, so as to establish unobviousness of the presently claimed invention, even where the teachings of the applied references were to establish a prima facie case of case of obviousness (as discussed infra, it is respectfully submitted that the applied references do not establish such prima facie case).

Moreover, it is respectfully submitted that the teachings of these applied references would have neither disclosed nor would have suggested such material as in the present claims, having the layer of carbon with the weight ratio of the layer of carbon to a respective graphite particle and other parameters as in the present claims, and wherein this layer “consists essentially of” (see claim 12), or “consists of” (see claim 13), carbon; and/or wherein the carbon covers the respective graphite particle (see claim 14).

In addition, it is respectfully submitted that the teachings of the applied references would have neither disclosed nor would have suggested such material as in the present claims, having the layer of carbon with weight ratio of layer of carbon to the respective graphite particle, and, additionally, wherein the material has features as in the remaining dependent claims, including (but not limited to) slurry viscosity as in claim 3; and/or bulk density and rate of variation of bulk density as in claim 4.

The present invention relates to material for a negative electrode of a nonaqueous electrolyte secondary battery, and the negative electrode and the secondary battery formed respectively using such material and such negative electrode. The nonaqueous electrolyte secondary battery formed using such electrode and material can suitably be used in portable electronic devices, electric automobiles, electricity storage or the like.

Graphite particles for negative electrode material, in which a plurality of flat particulate are assembled or bonded so that a plurality of alignment surfaces may be non-parallel with each other, thereby forming the graphite particles, have been proposed, as described in the paragraph bridging pages 3 and 4 of Applicants' specification. However, as described in the first four lines on page 4 of Applicants' specification, there is a problem that the charging capacity (charge load

characteristics), when the battery using such graphite particles is charged at a high speed, is low.

While, as described in the sole full paragraph on page 4 of Applicants' specification, it has been disclosed to coat a surface of a graphite particle with low crystalline carbon, the published application disclosing such coating does not mention any advantage in connection with charge load characteristics. When the carbon is simply coated on the surface of the graphite particles, without regard to amount, features of the use of non-aligned graphite particles are lost; that is, discharge capacity is reduced, and charge/discharge efficiency is deteriorated.

A further problem in connection with previously proposed negative electrode material utilizing graphite particles, even having a carbon coating, is that adhesiveness of the electrode material in the negative electrode is unsatisfactory. That is, the negative electrode material of the negative electrode tends to peel after pressing.

Against this background, Applicants provide negative electrode material for a nonaqueous electrolyte secondary battery, which has excellent discharge capacity, charge/discharge efficiency and charge load characteristics, and which has satisfactory adhesiveness. Applicants have found that by providing a layer of carbon on the surface of each of the graphite particles, which graphite particle has a structure where a plurality of flat graphite fine particulate assemblies or bonds non-parallel with each other, with a ratio (by weight ratio) of the layer of carbon to a respective graphite particle being in a range of 0.001-0.01, and having other properties as in present claim 1, including bulk density, specific surface area and R value, objectives according to the present invention are achieved. That is, as described in the paragraph bridging pages 8 and 9 of Applicants' specification, when the ratio of the carbon layer to graphite particle is less than 0.001, an improvement

width in the charge load characteristics is small; while when the ratio exceeds 0.01, the initial charge/discharge efficiency is deteriorated. By providing the weight ratio as in the present claims, charge load characteristics are excellent, with excellent initial charge/discharge efficiency.

In addition, the presently claimed subject matter includes graphite particles having a volume of fine pores in the range of  $10\text{-}10^5$  nm in a volume of 400-2000  $\text{cm}^3/\text{kg}$ . When the volume of pores in the range is less than 400  $\text{cm}^3/\text{kg}$ , the discharge load characteristics and the discharge capacity tend to decrease, while, on the other hand, when the volume exceeds 2000  $\text{cm}^3/\text{kg}$ , the cycle characteristics tend to deteriorate. Note the paragraph bridging pages 9 and 10 of Applicants' specification.

Moreover, as is described in paragraphs [0025]-[0027] on pages 13 and 14 of Applicants' specification, when a bulk density, specific surface area or R value is outside the scope of the present claims, adhesiveness of the electrode tends to deteriorate.

As to advantages achieved by the present invention, note also the paragraph bridging pages 21 and 22 of Applicants' specification.

In connection with advantages achieved according to the present invention, note also Table 4 on page 31 of Applicants' specification, particularly Examples 1-4 of the present invention as compared with Comparative Examples 2 and 3, respectively containing ratios greater than, and less than, the ratio range in the present claims. As stated in the first paragraph on page 32 of Applicants' specification, it can be seen that material according to the present invention is excellent in discharging capacity, charge/discharge efficiency and charge load characteristics.

In Examples and Comparative Examples in the above-identified application, a bulk density under pressure of 33 MPa and a rate of variation of the bulk density when the pressure is released, were measured. According to this measurement, in Comparative Examples 2 and 4, where a ratio of the layer of carbon to a respective graphite particle is greater than the upper limit of 0.01 recited in the present claims, the electrode tends to peel after pressing, since in Comparative Examples 2 and 4 the rate of variation of the bulk density when the pressure is released exceeds 0.3, as seen respectively in Tables 2 and 5 respectively on pages 29 and 34 of Applicants' specification. Note also Experiments 1 and 2 of the enclosed Declaration, especially the rate of variation of the bulk density in Table 1 thereof. As can be recognized therefrom, with values for weight ratio, as well as with other properties as in the present claims including, inter alia, bulk density, specific surface area and R value, adhesiveness of the electrode is also unexpectedly improved.

Attention is again directed to Table 1 of the enclosed Declaration, setting forth, in tabular form, results of Examples 1-4 and Comparative Examples 1-3 in Applicants' disclosure, as well as results of the two additional Experiments. These two additional experiments, together with Examples and Comparative Examples in Applicants' original disclosure, show that the superficial carbonization rate is to be in the range between 0.001 and 0.01 to obtain an excellent effect not only in the charge load characteristics but also in the bulk density variation rate when pressure is released.

In Item 3 on page 2 of the Office Action mailed April 28, 2010, the Examiner acknowledges that Tables 2 and 5 in Applicants' disclosure "appear to show that the bulk density variation scales with the superficial carbon rate" but goes on to state that if "this is due to peeling it would be expected that this would increase as the amount of superficial carbon is increased". This statement by the Examiner

respectfully challenged, moreover, it is respectfully submitted, that the Examiner has not shown expected results in both charge load characteristics” and “bulk density variation rate when pressure is released” for the specific weight ratio range as in the present claims. It is respectfully that this weight ratio range has unexpectedly better results, providing a basis for a conclusion of unobviousness of the presently claimed subject matter even were a prima facie case of obviousness to be established.

To summarize, it respectfully submitted that by providing graphite particles whose surface is coated with a layer of carbon, a ratio (by weight ratio) of the layer of carbon to a respective graphite particle being in the range of 0.001-0.01, unexpected advantages are achieved in that the covered graphite particle can maintain the characteristics of the non-aligned graphite particles, while having excellent discharge capacity, charge/discharge efficiency and charge load characteristics.

Takei, et al. discloses composite carbon particles containing a graphite part, an amorphous carbon part and silicon, the composite carbon particle being produced by mixing a graphitic particle with an organosilicon compound and a carbon precursor, heating the resultant mixture and decomposing and carbonizing the organosilicon compound and carbon precursor. Note the English language abstract of Takei, et al. Note also paragraphs [0011] and [0012] of this patent document, describing, inter alia, that the graphite particles are graphite particles in which flat-shaped particles gather or combine with non-parallel relationship.

As recognized by the Examiner, e.g., in the second full paragraph on page 5, of the Office Action dated April 28, 2010, Takei, et al. would have neither disclosed nor would have suggested such features of the present invention including, inter alia, the recited ratio (by weight ratio) of the layer of carbon to a respective graphite particle, or R value, as in the present claims.



Moreover, it is respectfully submitted that Takei, et al. would have neither disclosed, nor would have suggested, and in fact would have taught away from, such material as in the present claims, including the specified layer of carbon on the surface of each of the graphite particles, having the recited ratio of the layer of carbon to a respective graphite particle of 0.001 to 0.01, or other properties recited in claim 1, and wherein the layer consists essentially of carbon (see claim 12), or consists of carbon (see claim 13), as in various of the present claims. In this regard, it is noted that Takei, et al. requires silicon in the material covering the graphite grains.

It is respectfully submitted that the additional teachings of Kitagawa, et al. or Ishii, et al. would not have rectified the deficiencies of Takei, et al., such that the presently claimed invention as a whole would have been obvious to one of ordinary skill in the art.

Kitagawa, et al. discloses a carbon material for negative electrodes of a lithium ion secondary cell, which is a carbonaceous powder having a plural-layer structure with a surface layer of carbonaceous matter, the carbonaceous powder being prepared such that, using a lumpy graphite powder as nucleus, this nucleus or the graphite powder is covered with a carbon precursor, which is then fired in an inner gas atmosphere at a temperature within a range of 700-2800°C, causing the surface layer of the carbonaceous matter to form, wherein the lumpy graphite powder has specified characteristics (1)-(6) as set forth in paragraph [0012] on page 3 of this patent document. Note also, paragraph [0018] bridging pages 4 and 5 of this patent document, further describing the carbon precursor for covering the surface of the graphite particle nucleus; and note also paragraph [0019] on page 5 of this patent document, describing that the rate of the carbonaceous matter in the

carbonaceous powder of plural-layer structure is adjusted to be 0.1 wt % or more and not exceeding 50 wt % plural.

Initially, it is respectfully submitted that one of ordinary skill in the art concerned with in Takei, et al. would not have modified the teachings thereof in view of the teachings of Kitagawa, et al., without hindsight guidance from Applicants' disclosure, which hindsight guidance of course is improper under the guidelines of 35 U.S.C. 103. In this regard, attention is respectfully directed to Table 3 on page 13 of Kitagawa, et al. As can be seen therefrom, no change was observed in 1/5C discharge capacity, 1C discharge capacity and 1C discharge capacity at -20° C, as a result of using carbonaceous powder of a plural-layer structure. As to what is meant by 1/5C and 1C discharge capacity, note for example, paragraph [0056] bridging pages 13 and 14 of Kitagawa, et al.; note also paragraph [0049] on page 10 of this patent document. As can be seen from paragraph [0057] on page 14 of Kitagawa, et al., similar properties were obtained with a weight ratio of carbon coating to graphite particle within the range of 99:1 to 50:50. Thus, it is respectfully submitted that Kitagawa, et al. discloses that the discharge capacity does not change even where there is a carbon layer on the surface of the graphite particle. In view of the teachings of this reference, it is respectfully submitted that one of ordinary skill in the art would not have been guided to the specific range for the ratio (by weight) of carbon coating to graphite particle, as in the present claims, and the teachings of Kitagawa, et al., even in combination with teachings of Takei, et al., would not have disclosed or suggested the improvement in discharge capacity (charge load characteristics) by regulating the ratio of superficial carbon to graphite powder, much less the advantages achieved using the specific ratio (by weight) of the present claims.

Ishii, et al. discloses graphite particles for use in a negative electrode for lithium secondary batteries, the graphite particles being obtained by assembling or binding together a plurality of flat-shaped particles so that the planes of orientation do not become parallel to one another. Note, in particular, paragraphs [0013] and [0014] on page 1 of this patent publication. Note also paragraphs [0065] and [0071]-[0075] on page 5 of this patent document, disclosing that the graphite paste used in forming the negative electrode for the lithium secondary battery includes an organic binder, which organic binder can be polyethylene, polypropylene, ethylene-propylene terpolymer, butadiene rubber, styrene-butadiene rubber, butyl rubber, polymeric compounds having a high ionic conductivity, and the like.

Contrary to the contention by the Examiner, it is respectfully submitted that the combined teachings of Takei, et al., and Ishii, et al., would have neither disclosed nor would have suggested the presently claimed invention, including, inter alia, the ratio (by weight ratio) of the layer of carbon to a respective graphite particle, and with, inter alia, bulk density, specific surface area and R value, among other features as discussed previously.

The contention by the Examiner that Ishii, et al. would have disclosed or would have suggested, in combination with the teachings of Takei, et al., such ratio of a layer of carbon formed on the surface of each of the graphite particles, is respectfully traversed. As applied by the Examiner, Ishii, et al. discloses a binder. Ishii, et al. discloses that a mixing ratio between the graphite particles and the binder (not a layer of carbon formed on the surface of each of the carbon particles) is that recited in the reference, per 100 parts by weight of graphite particles. It is respectfully submitted that the binder in Ishii, et al. is used for bonding graphite particles with each other, and not for forming a layer on a surface of the graphite particle. It is respectfully submitted that such binder disclosed in Ishii, et al., would

have neither taught nor would have suggested the layer of carbon formed on the surface of the graphite particles as in the present claims, much less in the weight ratio as in the present claims, and advantages achieved thereby.

Thus, it is respectfully submitted that the binder in Ishii, et al., is a binder necessary to produce negative electrode material for a secondary battery, and is used to bond between a negative electrode material and a current collector. It is respectfully submitted that this binder material in Ishii, et al. is not for forming a layer of carbon on a surface of the graphite particles.

As Ishii, et al. discloses a binder material for, e.g., bonding the graphite particles to a current collector, amount of binder as disclosed in Ishii, et al, even together with the teachings of Takei, et al., would have neither taught nor would have suggested the weight ratio of carbon layer to graphite particle as in the present claims, and advantages thereof.

Thus, while the Examiner relies on the teachings of Ishii, et al. as disclosing the weight ratio in the present claims, as discussed in the paragraph bridging pages 5 and 6 of the Office Action dated October 14, 2009, in view of the purpose of the binder in Ishii, et al., including its purpose of binding to the current collector, it is respectfully submitted that the teachings of Ishii, et al., even in combination with the teachings of Takei, et al., would have neither disclosed nor would have suggested the presently claimed subject matter, including, inter alia, the weight ratio of the layer of carbon to the graphite particles, the layer being on the surface of the graphite particles, in particular covering the graphite particles (see claim 14), and advantages achieved thereby.

The unexpectedly better results in both bulk density variation rate and charge load characteristics, upon coating carbon in an amount (weight ratio) relative to the graphite particle, as in the present claims, is again noted. Such unexpectedly better

results can be seen from the evidence of record, including the enclosed Declaration. Such unexpectedly better results support a conclusion of unobviousness of the presently claimed invention.

In view of the foregoing comments, and in light of the evidence of record, reconsideration and allowance of all claims presently in the above-identified application are respectfully requested.

To the extent necessary, Applicants hereby petition for an extension of time under 37 CFR 1.136. Kindly charge any shortage of fees due in connection with the filing of this paper, including any extension of time fees, to the Deposit Account of

Antonelli, Terry, Stout & Kraus, LLP, Account No. 01-2135 (case 1204.46017X00),  
and please credit any overpayments to such Deposit Account.

Respectfully submitted,  
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